Probabilities of nonradiative transitions at acceptor centers

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A technique based on the general angular-momentum formalism is developed for the computation of the probabilities of quantum transitions at acceptor centers in crystals with a degenerate valence band. This technique is used to compute the probabilities of single-phonon nonradiative transitions between the levels of the discrete spectrum of the acceptor. It is shown that in the region of large values of the parameter qa the probabilities decrease rapidly with increasing qa (q is the phonon wave vector and a is the radius of the center) and that the transition to the ground state is a bottleneck in the intracenter relaxation process. As applied to the parameters of Ge, the computation of the lifetime of the first excited state of the acceptor leads to large values (up to roughly 10^{-7} sec), in accord with the experimental data.

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1. INTRODUCTION

In a wide class of semiconductors with a cubic lattice the valence band is degenerate. This makes the energy spectrum of shallow acceptor centers nontrivial and, in many respects, very interesting. After the original work by Kohn and Shechter,[1] quantitative acceptor-center calculations were performed with various degrees of accuracy by a number of authors.[2-5] Concomitantly, the optical spectra of such centers were investigated experimentally (for a review, see, example, Ref. 6). There is now clarity in respect to the classification of the acceptor levels, and an entirely satisfactory quantitative agreement between the theoretical and experimental data on the locations of the levels has been achieved.

Meanwhile, the problem of the computation of the intensities of the quantum transitions between acceptor levels has virtually not been formulated. Such computations are technically difficult, since, because of the multicomponent structure of the acceptor wave functions, the integration over the angular variables involves a large number of spherical harmonics with different angular momenta. These difficulties are especially great in the particular case of calculations of the probabilities of nonradiative transitions occuring in acceptors with the emission of phonons.

Little attention has on the whole been paid to single-phonon relaxation processes in shallow impurity centers. This is connected with the assumption usually made in the analysis of the rate of recombination processes in semiconductors in the spirit of the cascade theory that the bottleneck is the process of carrier capture from the band by one of the levels of the impurity center—generally speaking, not by the ground level, but by one of the excited levels. In this case the rate of relaxation of the excited states, i.e., the rate of transition of the captured carrier to the ground level, is assumed to be sufficiently high. As applied to shallow impurity centers, where the relaxation is a single-phonon process, this appeared to be a particularly reasonable assumption. From such a standpoint, the computation of the probabilities of intracenter relaxation processes was not essential; the rate of the recombination processes is determined by the rate of capture from the band by the impurity center, while the subsequent fast intracenter-relaxation processes virtually do not manifest themselves, and are difficult to observe.

However, recently, Gantmakher and Zverev,[7] in analyzing the experiments on the magnetic-impurity oscillations of the low-temperature photoconductivity of p-Ge, arrived at the conclusion that the relaxation time of the first excited state of acceptors in Ge is long: $\tau \sim 10^{-6} - 10^{-7}$ sec. The preliminary results of our computations[8] agree with $\tau \sim 10^{-7}$ sec. Close values, $\tau \approx 3 \times 10^{-7}$ sec (Ref. 9) and $\tau \approx 6 \times 10^{-8}$ sec, were also obtained in the latest experiments, which were performed by Gershenzon *et al.*,[10] using a completely different method.

A distinctive feature of the processes of relaxation to the ground state of shallow impurity centers is the extremely strong dependence of the transition probability on the parameters of the centers. Physically, it arises as a result of the following circumstance. The characteristic wave vector, q, of the phonons emitted in a transition to the ground state is given by the relation $E_i \sim \hbar s q$, where s is the velocity of sound and E_i is the ionization potential of the center; here account is taken of the fact that the first excitation potential of the center differs from E_i by not more than a factor close to two. Since E_i is connected with the radius, a, of the center by the relation $E_i \approx e^2/\varkappa a$ ($\varkappa$ is the permittivity),

$$qa = a/\lambda\!\!\!^{-} \approx e^2/\varkappa \hbar s \equiv \alpha^*; \qquad \text{(I)}$$

$\lambda\!\!\!^{-} = \lambda/2\pi$, λ being the phonon wavelength. For the usual values of the parameters in Ge-type crystals, the effective "fine-structure-constant" $\alpha^* \sim 30 \gg 1$. Therefore, we meet here a situation that is the opposite of what obtains in the optics of atoms and impurity centers, where expansions in terms of multipole moments are valid in intensity calculations. For $qa/2 \gg 1$, the transition matrix elements are determined largely by the behavior of the wave functions in the vicinity of the singularity of the potential, i.e., at small r, and decrease rapidly with increasing qa—not slower than $(qa)^{-4}$. For $qa/2 \ll 1$, the probabilities decrease rapidly with decreasing q.

The relaxation processes in shallow impurity centers

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were first considered by Ascarelli, Brown, and Rodriguez[11,12] within the framework of the hydrogenic model for a nondegenerate band in connection with the problem of recombination via such centers. It is clear from the foregoing that, as a result of the extremely rapid dependence of the probabilities on the parameters of the problem, attempts at a direct application of the obtained formulas to semiconductors with a complex band structure through the replacement of the Bohr radius by some effective "center radius" lead to a large uncertainty in the transition probabilities—up to several orders of magnitude. For example, because of the degeneracy of the valence band, the acceptor wave function contains two scales,[13] which are connected with the presence of two different masses (of the light and heavy holes); therefore, the concept of a center radius becomes ambiguous. In donors, the anisotropy of the effective mass has a significant effect on the probability values.[14]

At the same time, general qualitative laws are well conveyed by relatively simple formulas that are obtained for the hydrogenic model. For $T=0$, in the limit $qa/2 \gg 1$, we obtain for the probability of transition between the s levels n_1 and n_2 (it is assumed that $n_2 < n_1$) the expression[1)]

$$w_{n_2 n_1} = \frac{64 D^2 \hbar^4 s^3}{\pi \rho a^8 E_i^5} \frac{(n_1 n_2)^7}{(n_1^2 - n_2^2)^5}, \tag{IIa}$$

here D is the deformation potential, ρ is the density, s is the velocity of sound, and a is the Bohr radius. If we express the right-hand side of (IIa) in terms of qa, then

$$w_{n_2 n_1} = \frac{64 D^2}{\pi \hbar \rho s^2 a^3} \frac{1}{(n_1 n_2)^3 (qa)^5}, \quad qa = \frac{E_i a}{\hbar s}\left(\frac{1}{n_1^2} - \frac{1}{n_2^2}\right). \tag{IIb}$$

It can be seen from the formulas (II) that the dependence of the probability on the parameters is indeed a rapid one, and it also follows from (IIb) that, at fixed qa, the probability essentially depends on n_1 and n_2. The right-hand side of (IIa) monotonically depends on n_2 at fixed n_1; as a result, the transitions between neighboring levels predominate in the $qa/2 \gg 1$ case. On the other hand, at small qa values the probabilities decrease rapidly, and the relaxation at first proceeds in steps significantly exceeding the level spacing. At this stage it proceeds relatively rapidly, and then, when $qa/2 \gg 1$, it abruptly slows down.

The object of the present paper is to develop a technique for computing transition probabilities for acceptor centers. We focus our attention on the computation of nonradiative-transition probabilities. The consistent description of these transitions within the framework of the effective-mass method is based on the use of the matrix Hamiltonian of the electron-phonon interaction.[15] In the problem with degenerate bands great difficulties are presented by the integration over the angular variables. Therefore, we restrict ourselves to spherical bands, which allows us to use the standard techniques of angular-momentum theory, thereby significantly simplifying the calculations. At the same time, this approximation adequately exact, in any event, as applied to Ge.

The numerical computations confirm the existence of long intracenter relaxation times; this question is discussed in the final section of the paper.

2. THE BASIC EQUATIONS

In the spherical approximation the Hamiltonian of the acceptor has the form

$$H = \frac{\hbar^2}{2m}\left[\left(\gamma_1 + \frac{5}{2}\gamma\right)\mathbf{k}^2 - 2\gamma(\mathbf{kJ})^2\right] - \frac{e^2}{\varkappa r}, \tag{1}$$

where the J_α are the matrices of the moment $J = \frac{3}{2}$. Let us, following Baldereschi and Lipari,[5] choose the constant γ to be equal to $\gamma = (2\gamma_2 + 3\gamma_3)/5$; γ_1, γ_2, and γ_3 are the Luttinger parameters. The deformation-potential Hamiltonian describing the interaction of the hole with the acoustic phonons has, in the spherical approximation, the form

$$H_{el-ph} = D_{\alpha\beta}\varepsilon_{\alpha\beta} = \left(\mathfrak{a} + \frac{5}{4}\mathfrak{b}'\right)\varepsilon_{\alpha\alpha} - \mathfrak{b}'\varepsilon_{\alpha\beta}J_\alpha J_\beta, \tag{2}$$

where $\mathfrak{b}' = (2\mathfrak{b} + 3^{1/2}\mathfrak{d})/5$; $\mathfrak{a}$, $\mathfrak{b}$, and $\mathfrak{d}$ are the deformation potentials[15]; $\varepsilon_{\alpha\beta}$ is the strain tensor. The choice of $\mathfrak{b}'$ is made in much the same way as the choice of γ, and corresponds to the retention in H_{el-ph} of only the zeroth-rank spherical tensors; this definition of $\mathfrak{b}'$ is close to the one used by Bir *et al.*[16]

We shall also describe the vibrations of the crystal in the isotropic model. Then the acoustic phonons split into longitudinal and transverse phonons, and their spectrum is spherically symmetric. We shall not consider the interaction with the optical phonons, since their frequencies in Ge exceed $E_i/\hbar$.[17]

In the adopted model, the probability of transition of the hole from the state 1 into the state 2 with the emission of a phonon of the ν-th branch at low temperatures is equal to

$$W_{21\nu} = \frac{q^2}{8\pi^2 \rho u_\nu \Delta E} I_\nu, \quad I_\nu = \int |M(\nu, \mathbf{q})|^2 d\Omega_{\mathbf{q}},$$
$$M(\nu, \mathbf{q}) = \langle 2 | D_{\alpha\beta}\varepsilon_{\alpha\beta}(\nu, \mathbf{q}) e^{-i\mathbf{qr}} | 1 \rangle. \tag{3}$$

Here $\Delta E = E_1 - E_2$, $\mathbf{q}$ is the wave vector of the emitted phonon ($\hbar\omega_\nu(q) = \Delta E$), $u_\nu = d\omega_\nu/dq$ is the group velocity of the phonons,

$$\varepsilon_{\alpha\beta}(\nu, \mathbf{q}) = {}^1/_2[e_\alpha^\nu(\mathbf{q}) q_\beta + e_\beta^\nu(\mathbf{q}) q_\alpha], \tag{4}$$

and the $\mathbf{e}^\nu(\mathbf{q})$ are the unit polarization vectors. The integration over $\Omega\mathbf{q}$ in (3) is performed over the angular coordinates of the wave vector.

As a result of the presence of spherical symmetry and a center of inversion, the hole wave function can be represented in the form

$$\Psi_{nLFM}(\mathbf{r}, \sigma) = \sum_{lms} \langle lm, Js | FM \rangle R_{nLFl}(r) Y_{lm}(\Omega_r) \chi_s(\sigma), \tag{5}$$

where the Y_{lm} are spherical harmonics and the χ_s are the spin functions of the hole. The quantum number F determines the total angular momentum; M, its component; L is equal to $F - \frac{1}{2}$ or $F - \frac{3}{2}$; l assumes the values $l = L$, $L+2$; and n numbers of the levels with a given $\{LFM\}$ set. The factors $\langle lm, Js/FM\rangle$ are Clebsch-Gordan coefficients (CGC).

The computation I_ν integrals includes integration over the angular variables of $\mathbf{r}$ and $\mathbf{q}$, as well as over the

modulus of r. All the integrations over the angular variables can be performed in the general form; this will be done in the following section. On the other hand, the integration over r can be performed only approximately, since the functions R_{nLFl} are determined from numerical computations; we consider this problem in Sec. 4.

3. THE INTEGRATION OVER THE ANGULAR VARIABLES

The quantities I_ν [see the formula (3)] contain two independent integrations over angular variables: the integration in the matrix elements over Ω_r (the angular variables of the vector r) and the overall integration over Ω_q. Since, technically, this problem is rather tedious, we shall apply the tools of angular-momentum theory to the two integrations.

Let us first write the expression for $D_{\alpha\beta}\varepsilon_{\alpha\beta}$ [see (2)] in terms of irreducible spherical tensors. Expanding the tensor $\varepsilon_{\alpha\beta}$ and the "direct square"

$$(\mathbf{J}\otimes\mathbf{J})_{\alpha\beta}=J_\alpha J_\beta$$

in terms of irreducible tensors (Ref. 18, §107), we obtain

$$D_{\alpha\beta}e_{\alpha\beta}=\sum_{k=0,2}\sum_{\mu}A_k e_{(k)\mu}(\mathbf{J}\otimes\mathbf{J})^{\mu}_{(k)},\quad A_0=\frac{4}{5}\mathfrak{a},\quad A_2=-\mathfrak{b}'. \tag{6}$$

In the irreducible tensors, the subscript enclosed in brackets indicates the rank of the tensor. The index μ, which numbers the tensor components, is written as a subscript for covariant, and as a superscript for contravariant, components; for an arbitrary tensor, $T^{\mu}_{(k)}\equiv(-)^{k-\mu}T_{(k)-\mu}$. The absence in (6) of a term with $k=1$ is due to the fact that the tensor $\varepsilon_{\alpha\beta}$ is a symmetric tensor ($\varepsilon_{\alpha\beta}=\varepsilon_{\beta\alpha}$).

The tensor $\varepsilon(\nu,\mathbf{q})$, defined by the formula (4), can easily be rewritten in the form of a spherical tensor in the coordinate system, S_q, whose $0z'$ axis is oriented along q. Let us assign the index $\nu=0$ to the longitudinal polarization and, for the transverse phonons, choose as the $\mathbf{e}^\nu$ the complex unit vectors of circularly polarized waves and assign to ν the values $\nu=\pm1$. Then in the S_q system the components of the spherical tensors corresponding to the vectors q and $\mathbf{e}^\nu$ are equal to $\bar{q}_{(1)\mu}=iq\delta_{\mu 0}$, $\bar{e}^{\nu}_{(1)\mu}=i\delta_{\mu\nu}$. According to (4) and the general formula for the spherical-tensor product (Ref. 18, § 107), we have

$$\bar{\varepsilon}_{(k)\mu}=-q\langle 10,1\nu|k\mu\rangle \text{ for } k=0,2; \tag{7}$$

for the remaining k all the components $\bar{\varepsilon}_{(k)\mu}=0$. The components of the tensor $\varepsilon_{(k)}$ in the coordinate system, S, fixed to the crystallographic axes can be expressed in terms of the $\varepsilon_{(k)\mu}$ with the aid of finite-rotation matrices (i.e., Wigner's D functions; see, for example, Ref. 19, Ch. 4):

$$\varepsilon_{(k)\mu}(\nu,\mathbf{q})=-q\sum_{\mu'}\langle 10,1\nu|k\mu'\rangle D^{(k)}_{\mu'\mu}(\omega_q),\quad k=0,2, \tag{8}$$

where ω_q denotes the set of three angles determining the mutual orientation of the systems S and S_q. Incidentally, it follows directly from (7) that the interaction with the transverse phonons is determined solely by the deformation potential $\mathfrak{b}'$. Indeed, when $\nu=\pm1$, $\mu=\pm1$ also, and therefore the tensor $\bar{\varepsilon}_{(0)}=0$.

Similarly, expanding $e^{-i\mathbf{qr}}$ in terms of spherical waves in the system S_q (cf. Ref. 18, §34), and then going over to the system S, we obtain

$$e^{-i\mathbf{qr}}=\sum_{l=0}^{\infty}(4\pi)^{1/2}[l]j_l(qr)\sum_m Y_{lm}^*(\Omega_r)D^{(l)}_{0m}(\omega_q), \tag{9}$$

where $[l]\equiv(2l+1)^{1/2}$ and the j_l are spherical Bessel functions.

Now we can perform the integration over Ω_r in the matrix elements $M(\nu,\mathbf{q})$. The integrand in this integration contains three spherical harmonics arising from the wave functions of the states 1 and 2 and the expansion of the plane wave $e^{-i\mathbf{qr}}$ [see (5) and (9)]. The integral can be expressed in terms of the CGC (Ref. 18, §107):

$$\int Y^*_{l_3m_3}Y^*_{lm}Y_{l_1m_1}d\Omega_r=(-)^{(l_1-l-l_2)/2}\frac{[l][l_2]}{(4\pi)^{1/2}[l_1]}\langle l_20,l0|l_10\rangle\langle l_2m_2,lm|l_1m_1\rangle. \tag{10}$$

It follows from the Wigner-Eckart theorem that the matrix elements of the angular-momentum vector J entering into (6) can also be expressed in terms of CGC. The components of the spherical tensor corresponding to J are:

$$(J^{\mu}_{(1)})_{\sigma\sigma'}=-i[J(J+1)]^{1/2}\langle J\sigma,1\mu|J\sigma'\rangle. \tag{11}$$

Therefore, the components of the irreducible tensors $(\mathbf{J}\otimes\mathbf{J})_{(k)}$ can also be written in terms of CGC.

Thus, the matrix element $M(\nu,\mathbf{q})$ can be wholly expressed in terms of a sum of products of a few CGC and two D functions. For the subsequent integration over Ω_q, it is convenient to transform the product of the two D functions into a sum by expanding it in terms of D functions with different angular momenta (Ref. 19, Sec. 4.6):

$$D^{(k)}_{\mu'\mu}(\omega_q)D^{(l)}_{0m}(\omega_q)=\sum_j\langle l0,k\mu'|j\mu'\rangle D^{(j)}_{\mu'm+\mu}(\omega_q)\langle lm,k\mu|jm+\mu\rangle. \tag{12}$$

From the above-presented formulas we obtain the following expression for the transition matrix element:

$$\begin{aligned}M(\nu,\mathbf{q})=qJ(J+1)\sum_{l_1l_2k}(-)^{(l_1-l-l_2)/2}\frac{[l_2][l]^2}{[l_1]}A_k\mathscr{R}_{l_1l_2l}(q)\langle l_20,l0|l_10\rangle\\ \times\sum_j D^{(j)}_{\nu M_1-M_2}(\omega_q)\langle 10,1\nu|k\nu\rangle\langle l0,k\nu|j\nu\rangle\\ \times\sum\langle l_1m_1,Js_1|F_1M_1\rangle\langle l_2m_2,Js_2|F_2M_2\rangle\\ \times\langle Js_2,1\mu_2|Js\rangle\langle Js,1\mu_1|Js_1\rangle\langle 1\mu_1,1\mu_2|k\mu\rangle\\ \times\langle lm,k\mu|jM_1-M_2\rangle\langle l_2m_2,lm|l_1m_1\rangle,\end{aligned} \tag{13}$$

where

$$\mathscr{R}_{l_1l_2l}(q)=\int_0^{\infty}R_{n_2L_2F_2l_2}(r)R_{n_1L_1F_1l_1}(r)j_l(qr)r^2dr. \tag{14}$$

The last summation sign in (13) denotes summation over all the angular-momentum components that are encountered twice.

We carry out the transformation of this last sum,

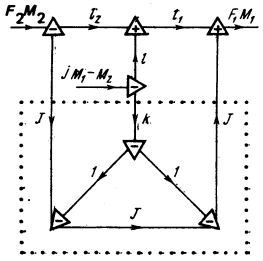
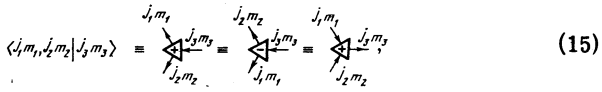


FIG. 1.

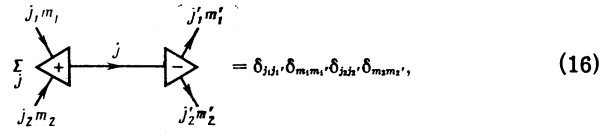
using Levinson's diagram technique[20] (see also Refs. 19, 21, and 22). We shall set the three-tailed vertices in correspondence with the Clebsch-Gordan coefficients according to the following rule:



(15)

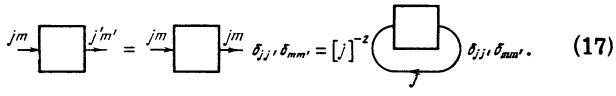
and with each angular-momentum component that enters into two CGC and over which a summation is performed, a line segment joining the corresponding vertices. Then the inner sum in (13) is represented by the diagram in Fig. 1. This diagram has three outward lines. Therefore, its dependence on the components of the angular momenta along the external lines should be the same as in the CGC $\langle F_2M_2, jM_1 - M_2/F_1M_1\rangle$; we shall compute the coefficient of proportionality below.

In transforming the diagrams, we use the orthogonality relation for the CGC, which, in the diagrammatic representation, has the form



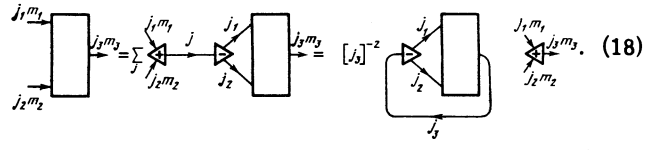
(16)

as well as the fact that any diagram with two external lines is diagonal in their quantum numbers and does not depend on the values of the angular-momentum component:



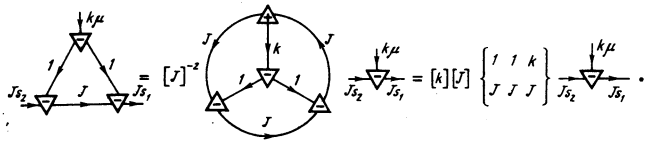
(17)

With the aid of these relations, an arbitrary three-tailed diagram can be transformed as follows:



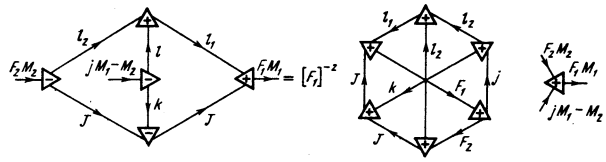
(18)

Let us now return to the diagram in Fig. 1. Separating out the part in it that is surrounded by points and transforming it with the aid of (18), we obtain



(19)

Here the closed four-vertex diagram has been expressed in terms of the 6*j* symbol. With the aid of (19), the diagram in Fig. 1 can be reduced to a simpler diagram, which, in its turn, can be transformed with the aid of (18):



$$=(-)^{l_1+l_2+l}[J][F_2][j][l_1]\begin{Bmatrix} l_1 & l_2 & l \\ J & J & k \\ F_1 & F_2 & j \end{Bmatrix}\langle F_2M_2, jM_1-M_2|F_1M_1\rangle; \quad (20)$$

the closed six-vertex diagram has been expressed in terms of the 9*j* symbol.

This completes the evaluation of the inner sum in (13), and we write out here the resulting expression for the matrix element:

$$M(\mathbf{v},\mathbf{q})=\sum_{jk} A_k T_{jk}(\mathbf{v},q)\langle F_2M_2, jM_1-M_2|F_1M_1\rangle [j] D^{(j)}_{\nu M_1-M_2}(\omega_q), \quad (21)$$

where

$$T_{jk}(\mathbf{v},q)=J(J+1)[J]^2[F_2][k]\langle 10,1\nu|k\nu\rangle \begin{Bmatrix} 1 & 1 & k \\ J & J & J \end{Bmatrix} \times\sum_{l_1l_2l}(-)^{(l_1-l-l_2)/2}[l_2][l]^2\langle l_20, l0|l_10\rangle \times\langle l0,k\nu|j\nu\rangle \begin{Bmatrix} l_1 & l_2 & l \\ J & J & k \\ F_1 & F_2 & j \end{Bmatrix} q\mathcal{R}_{l_1l_2l}(q). \quad (22)$$

In deriving (22), we took account of the fact that the factor $\langle l_20, l0|l_10\rangle$ is nonzero only for even l_1+l_2+l.

It now remains to carry out the integration over Ω_q in the formula (3). In order to use the orthogonality property of the *D* functions, let us go over from integration over Ω_q to integration over ω_q. This can be done, since the system S_q is specified up to a rotation about the vector **q**, and therefore one of the angles in the *D* functions is arbitrary; we can integrate over it and divide the result by 2π. As a result, we have

$$I_\nu=\frac{1}{2\pi}\int|M(\mathbf{v},\mathbf{q})|^2 d\omega_q=4\pi\sum_{jkk'}\langle F_2M_2, jM_1-M_2|F_1M_1\rangle^2 A_kA_{k'}T_{jk}T_{jk'}. \quad (23)$$

The entire dependence on M_1 and M_2 is contained in the square of the CGC. Normally, what is of interest is the transition probability summed over the mutually degenerated states belonging to the final level; after the summation over M_2, we obtain in place of (23) the expression

$$I_{\nu\ \mathrm{tot}}=4\pi\sum_{jkk'}A_kA_{k'}T_{jk}T_{jk'}. \quad (24)$$

Substituting (24) and (6) into (3), and separating out the deformation potentials $\mathfrak{a}$ and $\mathfrak{b}'$, we obtain for the probabilities for the transitions in which longitudinal phonons participate the expression

$$W_{2\to 1}=\mathcal{A}\mathfrak{a}^2+\mathcal{B}\mathfrak{a}\mathfrak{b}'+\mathcal{C}(\mathfrak{b}')^2 \quad (25a)$$

and for those in which transverse phonons participate the expression

$$W_{21t}=\mathscr{C}_t(b')^2. \tag{25b}$$

The computation of the coefficients $\mathscr{A}$, $\mathscr{B}$, $\mathscr{C}$, and $\mathscr{C}_t$ requires knowledge of the radial functions.

Everywhere above we assumed that the problem is spherically symmetric. However, we can obtain a generalization of the formula (23) with allowance for the splitting of the levels by the crystal field.

4. EVALUATION OF THE INTEGRALS INVOLVING THE RADIAL FUNCTIONS

To compute the matrix elements T_{jk}, we need to know the radial functions, which can be found only by numerical methods. We prefer a qualitative analysis allowing us to elucidate the main trends to a discussion of the numerical results.

As has already been noted in the Introduction, the hole transitions into the acceptor ground state (with energy transfer $\sim E_i$ to the phonon) occur when $qa \gg 1$, where a characterizes the dimension of the center. Therefore, here we give for the probabilities W the asymptotic formulas that follow from the asymptotic behavior of the matrix elements $\mathscr{R}$ [see (14)] at large q. It is, as usual, determined by the behavior of the integrand in the vicinity of the singularity, i.e., at small r. The investigation shows that, for $qa/2 \gg 1$,

$$W_{21v} \propto \frac{q^4}{(qa)^{2(L_1+L_2+4)}}, \tag{26}$$

i.e., W decreases not slower than $\propto q^{-4}$. In Ge the ground and first excited states are the states $1\Gamma_8^+$ and $1\Gamma_8^-$ (see Ref. 15),[2)] to which correspond the values $L=0$ and 1, respectively. Therefore, $W_{21v} \propto q^{-6}$. Of course, the formula (26) cannot be used for a quantitative comparison of the probabilities for the transitions between the various levels, since the coefficient that should be attached to the right member depends essentially on all the quantum numbers[3)]; this is clear even from the form of the formula (IIb). However, the formula (26) allows the comparison of the probabilities for the transitions that are accompanied by the emission of longitudinal (l) and transverse (t) phonons. With allowance for (3), we find that the probability $W \propto s^5$, and, as a result of the appreciable difference between the velocities ($s_l/s_t \approx 2$), it might be expected that the transverse phonons would not play a significant role in the first transition.

For transitions between close excited acceptor states with small qa,

$$W_{21v} \propto q^4 (qa)^{2(|L_1-L_2|-1|-1|}. \tag{27}$$

It is worth drawing attention to the fact that, as a result of the matrix structure of $H_{el\text{-}ph}$, here, in contrast to the case of optical spectra, monopole transitions with $L_1=L_2$ and $L_1=L_2\pm 2$ are possible; the intensities of the other transitions are of higher order in qa. At small q the intensities decrease not slower than $\propto q^4$. A comparison of (26) and (27) leads to the conclusion that the most intense should be the transitions in the intermediate qa range; in this respect, the picture is the same as in the case of nondegenerate bands (see the Introduction).

Let us now present some results of the numerical computations; more comprehensive data will be published separately. All the computations were carried out by us with ground- and excited-state radial functions, which were kindly made available to us by Doctor N. O. Lipari. These functions were given in the form of sums,

$$R(r)=r^s\sum_i Q_i \exp(-\alpha_i r), \tag{28}$$

containing up to twelve terms, the successive α_i values differing from each other by roughly a factor of two. The accuracy of these functions may be determined partly by the locations of the corresponding energy levels relative to the ground level $1\Gamma_8^+$. For the three lowest excited levels $1\Gamma_8^-$, $2\Gamma_8^+$, and $2\Gamma_8^-$, the corresponding values are equal to 5.89, 7.31, and 7.54 meV. The experimental level-energy values for a boron impurity in Ge are, according to Ref. 23, equal to 6.24, 7.57, and 7.94 meV; the discrepancies are apparently due mainly to the departure from the effective-mass method.

To illustrate the q dependence of the coefficients of the formulas (25a) and (25b), we show in Fig. 2 plots of $\mathscr{A}(q)$ and $\mathscr{C}(q)$ for two transitions. Plotted along the abscissa axis is the quantity $qa/2$, where a is the Bohr radius, determined from the mass $m=(m_l+m_h)/2$ (m_l and m_h are the light- and heavy-hole masses); it determines the characteristic scale for the variation of the wave function at small distances. The curves in Fig. 2 exhibit the general behavioral trends established above in the analytical investigation; they decrease rapidly as we move away from $qa/2 \sim 1$. They also show how strongly the transition probability depends on the location of the acceptor levels: a change in the energy by a factor of one and a half leads to roughly an order-of-magnitude change in the probabilities.[4)]

In Table I we give the values of the coefficients of the formulas (25a) and (25b) for the principal transitions

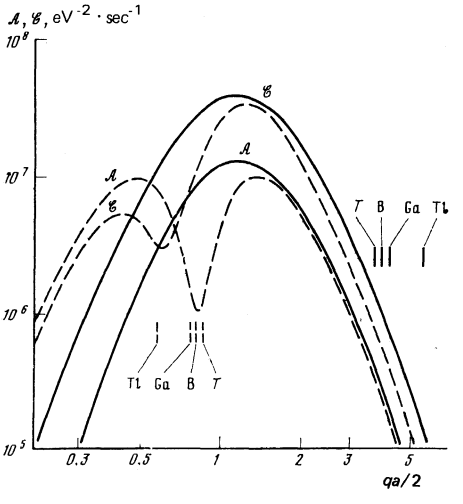


FIG. 2. Dependence of the coefficients $\mathscr{A}$ and $\mathscr{C}$ on qa for the transitions $1\Gamma_8^- \to 1\Gamma_8^+$ (the solid curves) and $2\Gamma_8^+ \to 1\Gamma_8^-$ (the dashed curves). Also indicated are the theoretical values (T) of the parameter qa (according to Lipari) and the experimental values of qa for several third-group impurities.

TABLE I.

Transition	Transition energy, meV	$\frac{q_l a}{2}$	$\mathscr{A}$	$\mathscr{B}$	$\mathscr{C}$	$\mathscr{C}_t$	Probability $W_{a=2}$ eV	Probability $W_{a=-5.6}$ eV
			$eV^{-2}\cdot sec^{-1}$					
$1\Gamma_8^- \longrightarrow 1\Gamma_8^+$	5.89	3.6	$5.99\cdot10^5$	$-1.78\cdot10^6$	$1.32\cdot10^6$	$2.17\cdot10^4$	$1.8\cdot10^7$	$2.7\cdot10^6$
$2\Gamma_8^+ \longrightarrow 1\Gamma_8^-$	1.42	0.87	$1.53\cdot10^6$	$-9.44\cdot10^6$	$1.71\cdot10^7$	$8.52\cdot10^6$	$2.0\cdot10^8$	$6.8\cdot10^7$
$2\Gamma_8^- \longrightarrow 2\Gamma_8^+$	0.23	0.14	$9.43\cdot10^5$	$8.22\cdot10^4$	$3.44\cdot10^5$	$1.26\cdot10^7$	$7.7\cdot10^7$	$1.0\cdot10^8$
$2\Gamma_8^- \longrightarrow 1\Gamma_8^-$	1.65	1.0	$9.76\cdot10^6$	$-1.90\cdot10^7$	$1.12\cdot10^7$	$1.86\cdot10^6$	$2.0\cdot10^8$	$1.2\cdot10^8$

between the four lowest levels; the remaining two transitions have a lower probability. Also given in the table are the theoretical energy values for the corresponding transitions and the values of the parameter $q_l a/2$ (for the longitudinal phonons). The q_l and q_t values were determined from the phonon dispersion curves.[17]

5. FINAL REMARKS

The parameters given in Table I allow us to find the transition probabilities from the formulas (25a) and (25b). Close values have been obtained for the deformation potentials b and b by various authors; we take $b = -2.2$ eV, $b = -4.4$ eV[24] and, consequently, $b' = -2.4$ eV. In contrast, the value of the "isotropic" deformation potential is not very reliably known. Two values $a = 2$ eV[25] and $a = -5.62$ eV[26] are given in the literature. We give the values of the probabilities $W = W_l + W_t$ for the corresponding transitions in the last columns of Table I. The probabilities change, on the average, by one order of magnitude, depending on the assumed value of a.

It can be seen from the table that the lifetime, τ, of the lowest excited state $1\Gamma_8^-$ is indeed long compared to the lifetimes of the higher excited states.[5)] For each of these states we can indicate at least one transition whose probability exceeds τ^{-1} by a factor of 10–100.

The uncertainty in the computed τ value arises largely as a result of the existing uncertainty in the value of the potential a.[6)] But the spread of the experimental values for τ (Refs. 7, 9, 10) is also considerable (see the Introduction). Therefore, we cannot give a particular preference to any of these values of a. Since the theory indicates the presence of a strong dependence of τ on the chemical shift (cf. Sec. 4), it would be interesting to measure τ for different impurities of the third group.

The obtained high τ values agree entirely with the magnetic-impurity-oscillation mechanism based on the inelastic scattering of electrons by excited acceptors.[7] But the question of the role of the competing mechanism[8] connected with the exciton decay on ionized acceptors remains, for the present, open.

The above-developed general method of computing probabilities can be applied in the theory of a number of other phenomena, e.g., in the theory of hypersound absorption by acceptors in deformed crystals and in magnetic fields, of the contribution of acceptors to thermal resistance as a result of resonance scattering of phonons on the split ground level of the acceptor, etc.

We are grateful to V. F. Gantmakher and V. N. Zverev for valuable discussions, to Dr. N. O. Lipari of IBM for making the acceptor wave functions available to us, and to L. N. Shchur for working out the program and performing the computer calculations.

[1)] The second factor in this formula differs from the one given in Ref. 11.

[2)] The number standing to the left of Γ is the ordinal number of the level with the given symmetry.

[3)] The complete asymptotic formulas for the probability of the first transition are derived in Ref. 8.

[4)] Of course, the short-range potential of the impurity leads not only to a chemical shift of the levels, but also to a change in the wave functions; however, the role of this factor is difficult to assess.

[5)] Since the smallness of the probabilities of the transitions to the ground state is connected with the fact that the factor qa is large for these transitions, the question arises whether the processes in which a large number of phonons participate could not compete with the single-phonon processes, on account of the fact that phonons with smaller qa values are emitted in these processes. However, the many-phonon transitions should have a smallness in powers of the electron-phonon coupling constant. Allowing for the smallness of this constant in Ge, as well for the fact that the probabilities differ by only one-two orders of magnitude, we find it difficult to expect the contribution of the many-phonon processes to be substantial.

[6)] Besides this, it is necessary to take account of the well-known difference[27] (see also Ref. 15, §32) between the values of the deformation potential under homogeneous-deformation and long-wave conditions; the second of them naturally enters into the transition matrix elements.

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Threshold instability and inhomogeneous states in nonequilibrium superconductors with optical and tunnel quasiparticle pumping

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A theory of the threshold instability in nonequilibrium superconductors with narrow quasiparticle sources (the electromagnetic field frequency, ω, and the voltage, V, across the junction satisfy the condition $\omega - 2\Delta \ll \Delta$, $V - 2\Delta \ll \Delta$) that was predicted in an earlier paper by the present author [Sov. Phys. JETP **39**, 862 (1974)] is developed. It is shown that, as a result of the development of the instability, a transition into an inhomogeneous state, which comprises regions with different finite (nonzero) order parameter values, is possible. The individual regions are separated by transition layers of width of the order of the quasiparticle diffusion length. In the case of a fixed voltage, V, across the junction (or of a given electromagnetic-field frequency ω), the inhomogeneous state is nonstationary. When the current through the junction is fixed (for a given level of absorption), the inhomogeneous state becomes stationary since the relative phase volume is fixed by the current. A broad range of experimental phenomena are well described by the theory.

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INTRODUCTION

It has been observed in a number of experimental investigations that nonequilibrium superconductors with optical and tunnel injection of quasiparticles go over into a new inhomogeneous state.[1-4] The nature of the inhomogeneous state of superconductors that are far from being in thermodynamic equilibrium has been intensively discussed in recent years. Two inhomogeneous-state models are known. The first model, first considered by Chang and Scalapino[5] and Baru and Sukhanov[6] (see also Ref. 7) is based on possible anomalous diffusion of nonequilibrium quasiparticles (the diffusion model). It is assumed that the diffusion proceeds from a region with a large value of the order parameter Δ (consequently, with a low quasiparticle concentration $\bar{n}$) into a region with a low Δ value (and a high $\bar{n}$), owing to the gradient $\partial\Delta/\partial r$. The instability condition in this model is extremely sensitive to the energy distribution of the nonequilibrium quasiparticles. Computations carried out with distribution functions satisfying a kinetic equation with a wide[8-10] and a narrow[11,12] quasiparticle source showed that the diffusion instability is apparently not realized under these conditions. This, of course, does not eliminate the possibility of realizing the diffusion instability in appropriate situations.

The second inhomogeneous-state model, which was proposed by the present author,[9] is based on the existence of a nonunique dependence of the order parameter on the quasiparticle-pump power β (or another corresponding parameter). In this model, the stratification of a homogeneous superconductor into regions of the normal ($\Delta = 0$) and superconducting ($\Delta \neq 0$) phases (or into regions with different values of Δ) can occur at a definite value, β_0, of β. For $\beta = \beta_0$, the energies of the phases with different Δ are equal, and the existence of a stationary phase boundary of width of the order of the quasiparticle-diffusion length or the coherence length is possible. This model is applied in Ref. 13 to nonequilibrium superconductors with tunnel injection [the superconductor-insulator-superconductor (SiS) junction].

According to Refs. 9 and 13, the inhomogeneous state is a nonstationary state for both optical pumping (fixed electromagnetic-field frequency) and tunnel injection in a fixed-voltage regime (fixed voltage, V, across the junction) with the exception of the $V = V_0$ regime. The phase boundary moves with a velocity proportional to $\beta - \beta_0$. If we neglect the time it takes the new phase to fill the sample, then the transition occurs discontinuously (first-order transition).

A stationary inhomogeneous state is attained in a regime in which a constant current flows through the

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